

AMENDMENTS TO THE SPECIFICATION

Please replace paragraph [0003] with the following amended paragraph:

[0003] Suitable ceramic membrane materials include mixed conducting, that is ion and electron conducting, metal oxides and dual phase combinations of ion conducting metal oxides and electron conducting metals and metal oxides. Exemplary ceramic compositions are disclosed in U.S. Patent Nos. 5,342,431; 5,599,383; 5,648,304; 5,702,999; 5,712,220; 5,733,435; 6,214,757; and Japanese Patent Application (Kokai) No. 61-21717. Ceramic membranes formed from an ion and electron conducting metal oxides typically exhibit the property of oxygen selectivity. "Oxygen selectivity" means that only oxygen ions are transported across the membrane with the exclusion of other elements and ions. Particular advantageous solid electrolyte ceramic membranes are made from inorganic oxides, typically containing calcium- or yttrium-stabilized zirconium or analogous oxides having a fluorite, brownmillerite or perovskite structure. Use of such membranes in gas purification applications is described in U.S. 5,733,069 5,733,435.

Please replace paragraph [0008] with the following amended paragraph:

[0008] The prior art has attempted to solve the problems associated with differential thermal expansion through use of internal metallic expansion joints and floating tubesheets. These devices are needed when sealing both ends of hot reforming tubes inside of reactor vessels. U.S. 5,567,398 teaches a compact steam reformer that utilizes multiple metallic bellows to accommodate differential thermal expansion of internal components. U.S. ~~5,567,933~~ 5,567,398 describes another reactor for steam reforming that specifically utilizes convective heat exchange between the product gas and the process feed gas. The heat exchanger tubesheet utilizes individual metallic bellows to accommodate differential thermal expansion. However, metallic bellows that can accommodate sufficient axial movement while operating at elevated temperatures often fail prematurely from fatigue and creep.

Please replace paragraph [0043] with the following amended paragraph:

[0043] Figure 10 is a horizontal cross-sectional view of the apparatus shown in Figure 1;

Please replace paragraph [0061] with the following amended paragraph:

[0061] Both exothermic partial oxidation reactions, as well as endothermic reforming reactions, will occur on anode side 29 of oxygen transport membrane tubes within the catalyst bed 28. The partial oxidation reaction for methane is shown in Equation 1. The steam reforming reaction for methane is shown in Equation 2. Additional conversion of carbon monoxide may occur with the exothermic water gas shift reaction, Equation 3.



Please replace paragraph [0062] with the following amended paragraph:

[0062] The scope of the present invention also includes reforming reactions between methane and CO₂ as indicated by Equation (4). The H₂/CO ratio in the syngas product can be adjusted somewhat by operating with various levels of steam-to-carbon ratios in the process feed. Similarly, the overall thermal balance between exothermic and endothermic reactions can be adjusted by altering the steam-to-carbon ratio.



Please replace paragraph [0068] with the following amended paragraph:

[0068] With reference again to Figure 2, a commercially-available nozzle-mix combustion burner 105 is used to provide heat to the reactor during startup. Air and natural gas are supplied to burner 105 which fires into a low pressure air

chamber directly below a tubesheet 106 that supports oxygen transport membranes 80. Heating rate for the reactor vessel 72 and internal components including the oxygen transport membrane tubes 80 is controlled by both the firing rate of the burner and the feed air flow rate. When steady state operating temperatures have been achieved and actual syngas production begins, the overall reaction will become exothermic and no longer require thermal input from the burner-104105.

Please replace paragraph [0073] with the following amended paragraph:

[0073] Referring to Figure 8, oxygen transport membrane tubes 80 will slide freely within reaction section 26 defined by catalyst bed 28 interspersed with optional porous shroud tubes 88. Shroud tubes 88 surround each oxygen transport membrane tube 80 (shown in Fig. 1) and form a means of gas communication between the oxygen transported through the ceramic membrane wall and the catalyst bed 28. The optional shroud tubes 88 also provide a means to remove and re-insert oxygen transport membrane tubes or remove and replace catalyst without catalyst bed-tube interaction. The catalyst bed 28 is supported from the fixed oxygen transport membrane tubesheet 106 below by means of supports 110 and can grow vertically upward as required from thermal expansion without constraint.

Please replace paragraph [0075] with the following amended paragraph:

[0075] Referring to Figure 9, a further internal component assembly that has freedom to grow from thermal expansion is first heat exchanger 20. First heat exchanger 20, which is supported within the reactor by fixing its tubesheet 116 between two reactor flanges 118, provides a means to recover heat from the hot oxygen depleted stream 18 exiting from the inside of the oxygen transport membrane tubes 80. First heat exchanger 20 is separated into stages that are divided by baffles 120122 which direct the shell side flow in a cross-counter flow fashion with respect to the tube side flow. Oxygen depleted stream 18 transfers heat to the incoming oxygen containing stream 10 as it flows through first heat

exchanger 20. As depicted in Figure 1, the oxygen containing stream 10 is delivered to the inside of the oxygen transport membrane tubes 80 by small diameter open-ended lance tubes 78 that extend internally to a position near their capped ends. The axial clearance between these open-ended lance tubes 78 and the inside surface of the cap of the oxygen transport membrane tubes 80 will accommodate the thermal growth of first heat exchanger 20 vertically upward.

Please replace paragraph [0078] with the following amended paragraph:

[0078] Figure 11 depicts a means to limit this undesirable communication by isolating the fuel and air streams with an intermediate buffer gas. The buffer gas should not support an oxidation reaction with fuel. Gases such as nitrogen, carbon dioxide, or steam would be acceptable candidates with preference depending on seal material selection. Detailed description of suitable seal designs is given in U.S. 6,139,810, Figures 3 and 4.

Sealing is accomplished in two stages with a buffer gas between seals 126 at tubesheet 106 and 128 at plate 127 which separates the buffer zone 130 from the reaction products. The buffer zone or chamber 130 adjacent to the seal 126 would be filled with the buffer gas through passageway 132 and maintained at a pressure slightly higher than the fuel stream. Leakage of this buffer gas into the high pressure fuel stream could be controlled by minimizing the pressure differential and/or utilizing a second set of mechanical seals for seal 128 around the oxygen transport membrane tubes 80. If steam and/or CO₂ are used as a buffer gas, a small amount of leakage is inconsequential and can be tolerated, since these gases are also constituents of the reacting stream. In this case employing a flow restriction in the form of close circumferential clearances between the OTM tubes and the openings in plate 127 or non contact labyrinth seals can be adequate for limiting leakage to permissible levels. Buffer gas leakage into the air stream will depend on the quality of seal 126 between the oxygen transport membrane tube 80 and the tubesheet 106.

Please replace paragraph [0082] with the following amended paragraph:

[0082] With reference again to Figure 2, when reactant stream 38 includes higher hydrocarbons other than methane, it is advantageous to include a pre-reforming process step to avoid operational problems resulting from carbon formation or coking. To such end a pre-reformer consisting of a catalyst bed 134 is integrated with Apparatus 1 in a way such that ~~first~~second heat exchanger ~~2039~~ is split into two separate sections 136 and 138 with one on each side of catalyst bed 134. The first section 136 preheats ~~to the oxygen-containing gas stream 10~~reactant gas 38 to raise its temperature up to the level normally associated with pre-reformers, about 450°C to about 550°C. The second section 138 provides the final heat exchange to bring the pre-reformed gas up to a temperature range of up to about 700°C and about 1000°C.

Please replace paragraph [0083] with the following amended paragraph:

[0083] Figure 12 depicts an optional annular jacket 140 for recovering additional heat from Apparatus 1 that would otherwise be part of the environmental heat leak. A portion of the reaction vessel wall 73 can be jacketed with a thin metallic skin 142 to form an annular flow passage 144 for preheating the oxygen containing stream 10. The preheated oxygen containing stream 10 can then be directed, via passageway 146 into the first heat exchanger 10 for further heating. This jacketed approach requires that the internal insulation thickness "t" of this portion of the pressure vessel be reduced slightly to raise the wall temperature and enhance the temperature difference and improve heat transfer. The temperature of the reaction vessel wall 73 must not rise to a level that reduces the corresponding material strength below what is required to safely contain the internal reactor pressure.